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Dependence of flame propagation on pressure and pressurizing gas for an Al/CuO nanoscale thermite

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Abstract

The pressure dependence of flame propagation in an Al/CuO nanoscale thermite was studied. Experiments were performed by loosely packing the Al/CuO mixture in an instrumented burn tube, which was placed in a large volume, constant pressure chamber with optical windows. A high-speed camera was used to take photographic data, and six pressure transducers equally spaced along the length of the burn tube were used to measure the local transient pressure. Ambient pressures were varied between 0 and 15 MPa, and three different pressurizing gases were used: argon, helium, and nitrogen. Three modes of propagation were observed. The pressure at which the mode of propagation changed was similar for argon and nitrogen, however, when pressurized with helium, transition occurred at lower pressures. In the low-pressure regime (~ 0 –2 MPa) a constant velocity mode with speeds on the order of 1000 m/s was observed. In this region, a convective mode of propagation was dominant. An accelerating regime was observed for a pressure range of approximately 2–5 MPa in argon and nitrogen, with speeds ranging from 100 to 800 m/s. In helium, however, if an accelerating region existed it occurred over a narrow pressure range which was not observed in the present experiments. An oscillating regime was observed in all three gases, in a pressure range of ~ 5 –9 MPa for argon and nitrogen, and a range of ~ 2 –4 MPa for helium. Velocities in this region are bimodal, and differ by orders of magnitude, suggesting that the propagation mechanism was oscillating between convective and conductive. At relatively high ambient pressures, a constant velocity mode with speeds on the order of 1 m/s was observed for all three gases. The conductive mode of propagation was likely dominant in this region.

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Keywords: Nanoaluminum; Thermite; Pressure dependence; Convective burning

1. Introduction

The dependence of the burning rate of solid energetic materials on the ambient pressure is of great practical value. Two important applications of solid energetics are for rockets and guns, both

of which operate at extremely high pressure and temperature [1]. For typical solid rocket propellants, the burning rate increases with an increase in pressure. A general explanation for this phenomenon is that an increase in pressure decreases the flame standoff distance, which increases the heat flow to the surface and consequently the regression rate of the surface [2]. For nitrocellulose or nitroglycerin double-base propellants, a pressure dependence of $r_b \sim P^n$ is exhibited, where

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n is a positive value normally between 0.7 and 0.9 [3]. In these systems, the majority of products from the combustion are gases, even at extremely high pressures. Also, these systems typically are not porous, so the hot gases formed by the reaction cannot penetrate forward through the propellant.

Another energetic material receiving interest lately is nano-thermites. Thermite reactions, which occur between a metal and a metal oxide, have relatively high energy densities. The combustion of thermites has long been of interest for various applications including explosives and high temperature combustion synthesis [4]. Classical thermite mixtures utilize micron-sized particles as their constituents. Typically, these materials have relatively high flame temperatures and propagation rates on the order of centimeters per second due to their relatively high packing densities and large particle sizes increasing the characteristic length scales for diffusion and mixing [5].

Reactions with loosely packed nanoscale thermites, however, have propagation rates on the order of 1 km/s in confined combustion experiments performed in tubes, and on the order of several hundred m/s in an open burn tray [6,7]. The smaller diameter particles decrease the time scales for mass diffusion and heat transfer. Moreover, the high surface area to volume ratio creates more contact points between fuel and oxidizer particles. This leads to a system of higher reactivity that demonstrates relatively fast propagation rates. For most thermite systems, a significant portion of the products ultimately end up in the condensed phase, even at atmospheric pressure. Furthermore, since they are comprised of particles, thermites are typically porous, which allows for convective burning.

Previous studies have shown various levels of pressure dependence on the burning rates for different formulations of classical thermites. Studies conducted by Maksimov et al. [8] on the Fe_2O_3 –Al– Al_2O_3 thermite mixture show that the reaction proceeds without gas evolution and is, therefore, independent of pressure between 1 and 100 atm. However, Ivanov et al. [9] have shown that Zr– BaO_2 , Mg– MoO_3 , and Zn– PbO_2 react with gas evolution and that the rates of reaction vary with pressure when pressed to relative densities of 0.6–0.8. An increase in propagation rate was observed up to some maximum point (between 1 and 5 MPa), which was followed by a decrease with further increase in pressure.

The propagation mechanism for nanoscale thermites is not fully understood. In order for the reaction to sustain propagation, energy must be transferred ahead of the reaction front. In porous mixtures, convection can play a key role in the propagation of the combustion wave. High pressures in the reaction zone create a pressure gradient, which allows for deep penetration of hot

gases into the interstitial volume ahead of the flame zone, heating the cold reactants. This phenomenon has been studied and shown to play a role in deflagration to detonation transition (DDT) [10,11] of typical solid propellant grains.

Previous work with loosely packed nanoscale thermites ($\rho \sim 5$ –10% TMD) suggests that the dominant propagation mechanism is convection [6,7]. Therefore, the amount of gas formed by the reaction plays a crucial role in flame propagation, since it creates the pressure gradient within the granular bed needed to drive the convective wave. Previous study was performed [12] whereby a nanoscale thermite was diluted with Al_2O_3 nanoparticles. The presence of these particles created lower flame temperatures, which significantly decreased the gas production and thus the propagation speed. In fact, the Al/CuO thermite eventually showed an unstable spiraling combustion wave when 20% by mass of Al_2O_3 was added to the system. Varying the ambient pressure could also decrease gas production, and thus hinder the convective propagation mechanism.

There has been no previous investigation of the pressure dependence of thermites with particles at the nanoscale. In contrast to the slower reactions of the classical thermites, nanoscale systems do not have sufficient time to allow for heat loss or depressurization of the system. In fact, for the fastest propagating mixtures, pressures at the reaction front attain values typical of a constant volume process [12].

In the present study, the combustion and reaction propagation of an Al/CuO thermite is studied with nanoscale ingredients at various ambient pressures to obtain a better understanding of the propagation mechanism. Being that the nano-thermites are largely comprised of interstitial volume, various pressurization gases (including He, Ar, and N_2) are studied.

2. Experimental

For this work, the particles were mixed by sonication in the same method as [6,12,13]. The aluminum particles were obtained from Novacentrix and have an average particle size of 38 nm. The copper oxide particles were from Sigma–Aldrich and have an average particle size of 33 nm. Both the aluminum and copper oxide particles were approximately spherical. Particle size and shape information was provided by the suppliers. The equivalence ratio of the mixtures was slightly fuel rich ($\Phi = 1.1$), which was found to be optimal by Sanders et al. [13]. The aluminum particles were determined to be 49% active aluminum by weight using a TGA analysis. To measure the propagation velocity, the burn tube experiment originally designed and used by Bockmon et al. [6] was used in this study with some modifi-

cations. An acrylic tube with length of 8.9 cm, inner diameter of 0.32 cm, and outer diameter of 0.64 cm, was placed in a polycarbonate block, which is shown in Fig. 1. Local pressure measurements along the length of the tube were obtained with dynamic pressure transducers (PCB 111A22/062A01) at six locations 1 cm apart. The pressure transducer ports were filled with O-ring lubricant to thermally insulate the piezo-crystals. The acrylic tube was filled with approximately 300 mg of the thermite material creating a packing density of 6.6% TMD.

The block was mounted vertically inside of an optical pressure vessel with a volume of 23 L. The vessel, also shown in Fig. 1, is described in detail in [14]. A Phantom v7.3 high-speed camera was used to take photographic images for calculating the burning rate from trajectory plots of the reaction front position versus time. The mixture was ignited using a resistively heated (15 V, 5 A) nichrome wire, which was embedded in the top of the nano-thermite mixture.

3. Results

3.1. Equilibrium calculations

Analysis of the equilibrium temperature and species composition of the Al/CuO system provides some insight into the phenomenon of the pressure dependence of combustion propagation. Equilibrium calculations were performed using CHEETAH 4.0 [15] with the JCZS product library developed by Hobbs et al. [16]. Figure 2 shows the equilibrium temperature, total gas phase mole fraction, and total condensed phase mole fraction at equilibrium with a constant volume system. Also plotted is the vaporization temperature of aluminum versus pressure. Constant pressure calculations were also performed, but constant volume calculations have been found more applicable for high velocity propa-

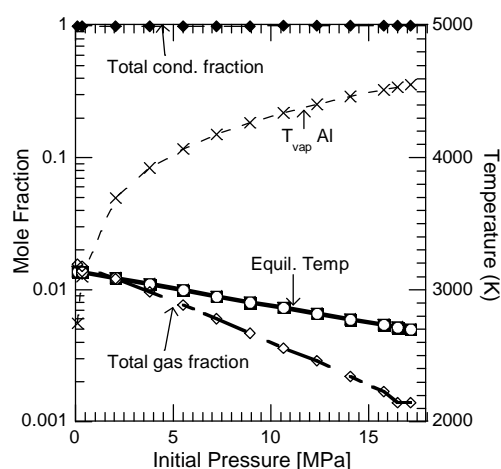


Fig. 2. Constant volume equilibrium data for the reaction of Al and CuO in argon. The mixture equivalence ratio was 1.1 and the initial temperature was 300 K.

gations, since the reactions are fast and will not allow time for depressurization within the reaction zone. For example, Malchi et al. [12] found reasonably good correlation between constant volume calculations and experimentally measured values for peak pressure with the Al/CuO nano-thermite. These calculations included both the Al_2O_3 and argon in the reactants. The Al_2O_3 is included due to the oxide shell, which is a significant percentage of the overall weight for the 38 nm aluminum particles used in this study (51% aluminum oxide, by mass). The amount of argon in the system for a given pressure was calculated according to the perfect gas law, holding the amount of interstitial volume constant. For the constant volume calculations in Fig. 2, the equilibrium temperature decreased with increasing ambient pressure for the entire range examined (0.1–18 MPa). This result is attributed to the increase in the amount of argon present in the interstitial sites of the mixture. As the amount of argon in the system is increased, more of the energy released from the exothermic reaction is used in heating the inert argon. Only small portions of the equilibrium products are in the gas phase for the constant volume system. Equilibrium calculations showed similar results when performed using helium and nitrogen as the interstitial gases. At high pressures, the N_2 system showed a significant reduction in the flame temperature (8% reduction from the argon equilibrium temperature at 15.8 MPa), due to the higher specific heat. Although the possibility exists for reactions between the nitrogen and the aluminum or oxygen in the system, the mole fractions for the products of such reactions were found to

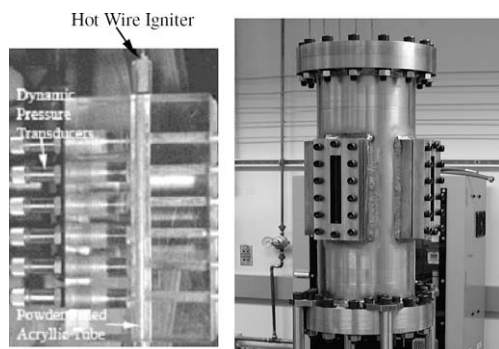


Fig. 1. Instrumented polycarbonate block and 23-L optical pressure vessel.

be insignificant at equilibrium. The equilibrium temperatures for different pressurizing gases are shown in Fig. 3.

3.2. Burning rate experiments

Figure 4 is an example of the visible emission data acquired using the Phantom v7.3 high-speed digital camera. These images were used to find the position of the flame front as a function of time. Trajectory plots were created, as shown in Fig. 5. A linear trend line was fit to the constant velocity region of these plots, and the slope was used as the average velocity. For the results shown in Fig. 5, the non-linearities appear to be primarily at the beginning and end of the tube, which could be attributed to ignition and end effects. However, for trajectories such as those observed in Fig. 6, it is clearly shown that the velocity is not constant, and the flame front is accelerating. Subsequent experiments at higher ambient argon pressures showed a transition to an unsteady propagation of the flame front, as shown in Fig. 7. Unstable combustion in thermites has been observed in two characteristic modes in classical thermites, spinning and oscillatory [17]. Malchi et al. observed the former in previous studies

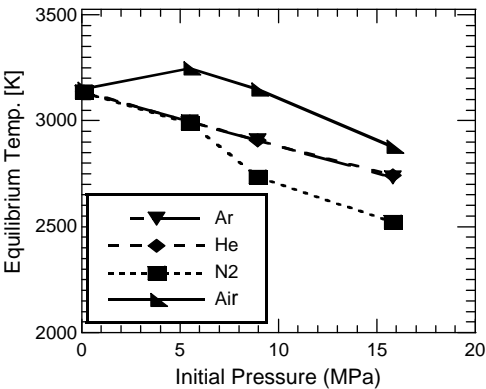


Fig. 3. Equilibrium temperatures for Al/CuO versus pressure for different pressurizing gases.

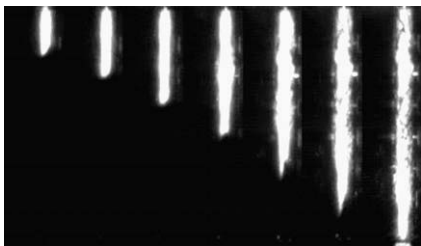


Fig. 4. Al/CuO thermite propagation in atmospheric air (frame interval: 13.5 μ s, equivalence ratio is 1.1).

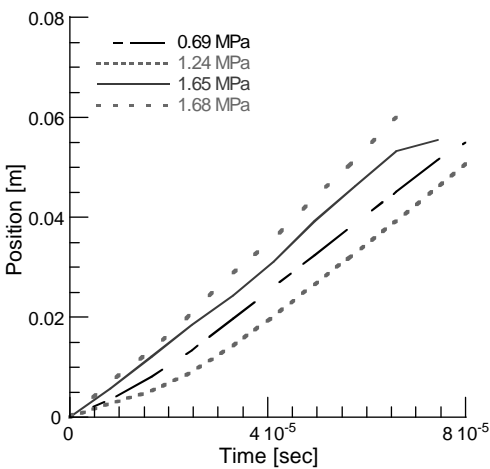


Fig. 5. Trajectory plots for Al/CuO nano-thermite at different ambient pressures in argon.

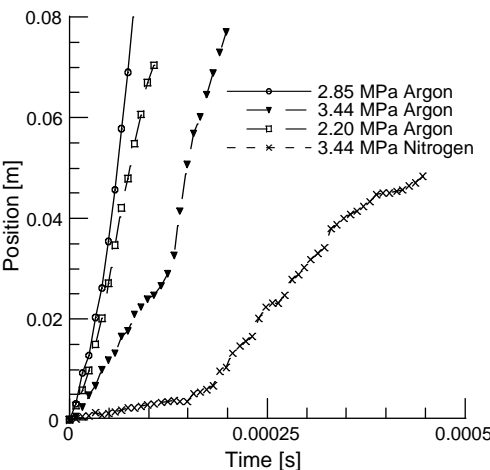


Fig. 6. Accelerating trajectory plots for Al/CuO at different ambient pressures in argon.

when the thermite was diluted with Al_2O_3 [12]. The instabilities observed here appear to be of the latter type.

Figure 8 shows the average combustion velocity as a function of the ambient pressure for experiments conducted with argon as the pressurizing gas. In the low ambient pressure region, between 0 and 2.9 MPa, the combustion velocity was approximately constant and on the order of 800 m/s. Accelerating propagation was observed over a small range of ambient pressures, between 2.9 and 3.4 MPa. An unstable, oscillating propagation was observed over a relatively wide range of pressures, from 4.2 to 8.9 MPa. At ambient pressures above 8.9 MPa, a slow, constant propagation was observed, with velocities on the order of 2 m/s.

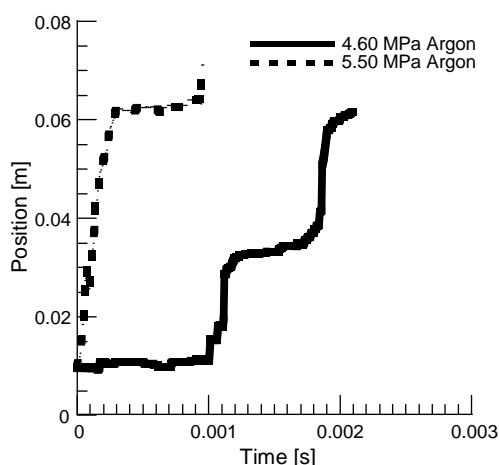


Fig. 7. Oscillating trajectory plots for Al/CuO at different ambient pressures.

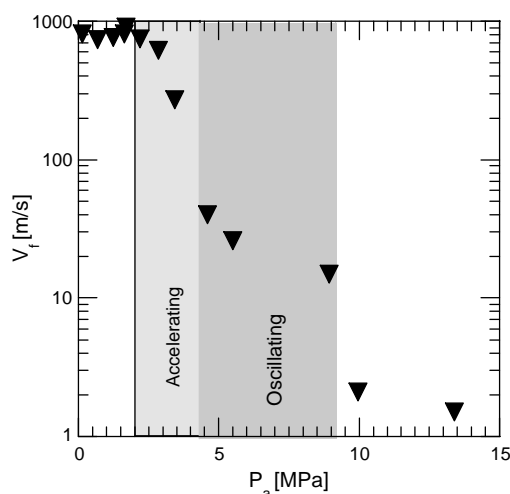


Fig. 8. Combustion velocity as a function of ambient pressure with argon as the pressurizing gas.

Experiments were also conducted using helium as the pressurizing gas. According to the CHEETAH calculations, there should be no difference between the equilibrium states when pressurizing with either argon or helium. Both gases are inert, monatomic and do not play a significant role in the chemical reactions. However, as shown in Fig. 9, the helium system transitioned directly to an oscillatory mode of propagation without any acceleration regime. This, as well as the transition to a slow constant propagation, occurred at lower ambient pressures than the system pressurized with argon. As a result, the ambient pressure range over which unstable, oscillating propagation was observed is significantly decreased. Since the equilibrium calculations

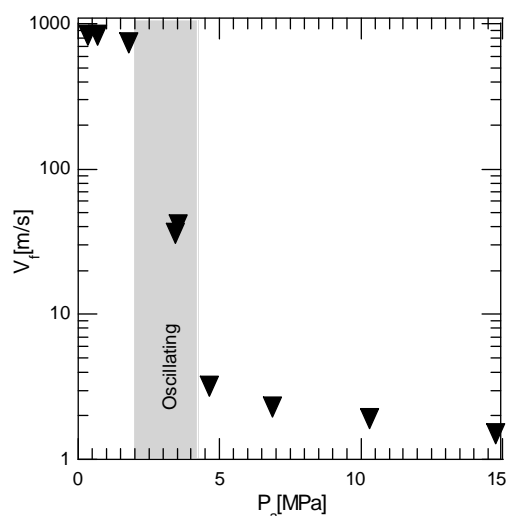


Fig. 9. Combustion velocity as a function of ambient pressure with helium as the pressurizing gas.

offer no insight into this result, it was determined that the disparity between the results observed must be related to the dynamic heat transfer properties of the respective gases. Helium has a very high thermal conductivity and thermal diffusivity, when compared with that of argon (Table 1) [18].

Experiments using nitrogen gas were performed in order to further investigate the change in transition point between the two inert gases. Nitrogen gas was selected because its thermal diffusivity is similar to that of argon. The specific heat, however, is greater, which was shown to reduce the flame temperature as shown in the equilibrium calculations. However, at mid-range ambient pressures, where the transition point was found to occur, the discrepancy in equilibrium temperatures was not significant (0.3% at 5.5 MPa, see Fig. 3). Also, as mentioned earlier, the nitrogen may react with the system, but the equilibrium calculations indicate the nitrogen containing products would be an insignificant percentage of the total products.

The combustion velocity of the Al/CuO nano-scale thermite in nitrogen gas is shown in Fig. 10 as a function of the ambient pressure. For an ambient pressure of 3.44 MPa, propagation of

Table 1
Thermal properties of pressurizing gases

Pressurizing gas	Thermal conductivity (mW/m K)	Specific heat (J/mol K)	Thermal diffusivity (cm ² /s)
Argon	17.9	20.8	192.7
Nitrogen	25.9	29.1	199.2
Helium	156.7	20.8	1683.5

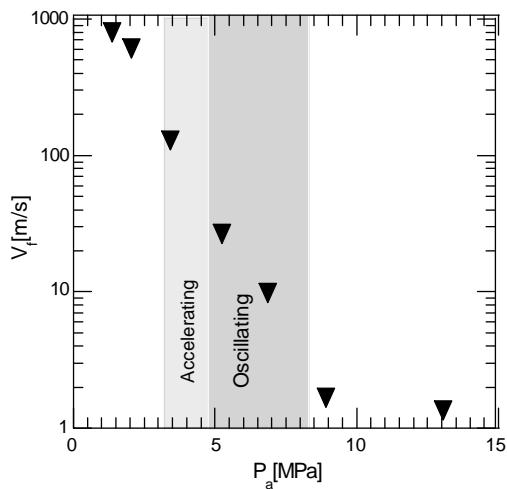


Fig. 10. Combustion velocity as a function of ambient pressure with nitrogen as the pressurizing gas.

the flame front was found to be accelerating. For experiments with ambient pressures of 4.6 and 6.9 MPa, the propagation was found to be in the unstable, oscillating regime. For runs at 8.9 and 13.4 MPa, the propagation was in the slow-constant velocity regime. The transition points to the unstable and slow-constant propagation regimes are found to match reasonably well with that of the argon system.

There exist many formal definitions for convective burning, as discussed by Asay et al. [11]. In general, convective burning is the heating of the energetic material up to the ignition point by the penetration of hot gases through the pores of the cold, un-reacted material, as caused by the presence of a pressure gradient. To quantify the effect of convective burning in the propagation of combustion, the local pressure was measured. Figures 11 and 12 represent two examples of pres-

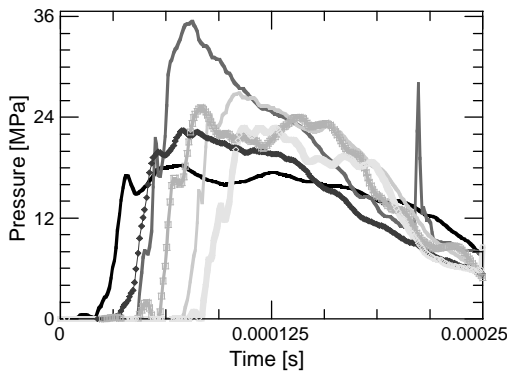


Fig. 11. Pressure overshoot for ambient pressure of 1.65 MPa in argon.

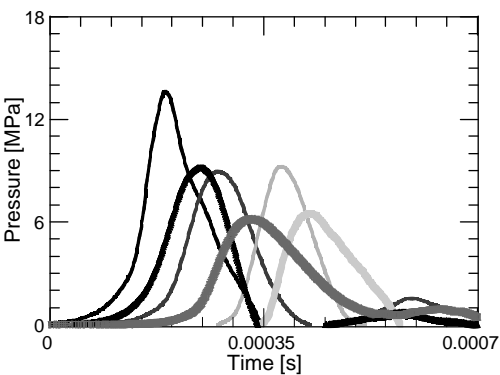


Fig. 12. Pressure overshoot for ambient pressure of 8.93 MPa in argon.

sure traces of the Al/CuO nanoscale thermite at ambient argon pressures of 1.7 and 8.9 MPa. The six different lines are the pressure traces for the six different pressure transducers. For the 1.7 MPa case, the pressure overshoot is relatively high compared to the ambient pressure, with an average of 21.5 MPa, about 13 times the ambient pressure. This reaction is also relatively fast, as can be seen by observing the time scale of the graph. For the 8.9 MPa case the average pressure overshoot is approximately 9 MPa, about the same as the ambient pressure.

The average pressure overshoot versus the ambient pressure is plotted for the argon, helium, and nitrogen systems in Figs. 13–15. The average value was obtained by taking the mean value of the peak pressure for each of the six pressure transducers. In the regions of fast-constant velocity propagation, the average pressure overshoot

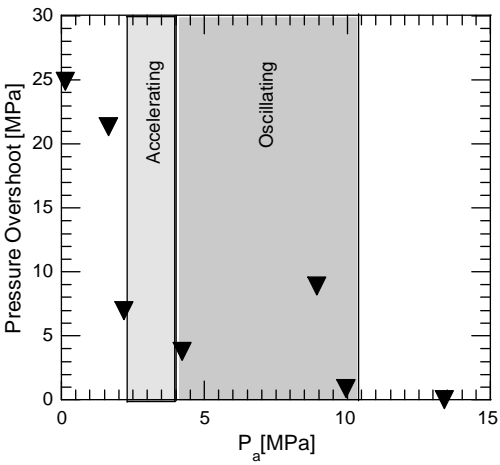


Fig. 13. Average pressure overshoot versus ambient pressure in argon.

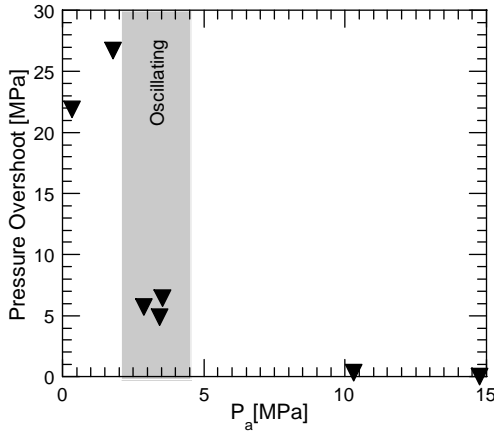


Fig. 14. Average pressure overshoot versus ambient pressure in helium.

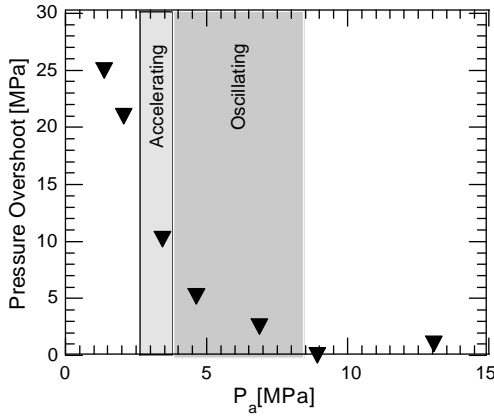


Fig. 15. Average pressure overshoot versus ambient pressure in nitrogen.

was found to be roughly 20 MPa on average. In the accelerating and oscillating regimes, the pressure overshoot was found to range between 4 and 10 MPa. In the slow-constant velocity regime, the pressure overshoot was found to range between 0 and 1 MPa.

4. Discussion and conclusions

The high propagation speeds and pressure overshoots observed in the low ambient pressure region confirms previous data, supporting the argument that the propagation mechanism is convection. The slow propagation speeds and low or zero pressure overshoot observed in the high ambient pressure region is evidence that convective burning does not play a role, and suggests the propagation mechanism is conduction. This means of propagation is similar to that of classical

thermites, with particle sizes on the micron scale. The oscillating and accelerating regimes of flame propagation suggests that for this range of pressures, convective and conductive heat transfer may both be important, and that the unsteadiness that is observed is an oscillation between convective and conductive propagation. To understand the difference in transition point for the different pressurizing gases, a ratio between convective and conductive heat transfer is created. For convective heat transfer, the heat flux to the unreacted, cold nano-thermite in front of the flame front can be represented by Eq. (1), where T_f is the flame temperature, T_0 is the initial temperature, v_g is the velocity of the hot gases, and ρ_g and $C_{p,g}$ are the density and specific heat of the hot gases. The heat transfer by conduction is represented by Eq. (2), where l_p is the length scale of the reaction zone, and k_{MIC} is the thermal conductivity of the nanoscale thermite

$$q''_{conv} = \rho_g C_{p,g} v_g (T_f - T_0), \quad (1)$$

$$q''_{cond} = k_{MIC} \frac{(T_f - T_0)}{l_p}. \quad (2)$$

Darcy's law is an empirical relationship used for modeling flow through a porous medium. If Darcy's law for flow through a particle bed is evoked [19], then v_g can be represented as a function of the pressure gradient, as in Eq. (3), where κ is permeability, ϕ is the porosity, and μ is the viscosity of the hot gas. The molar density of the hot gases expelled from the flame zone can be represented using the ideal gas law as shown in Eq. (4), where P_c is the ambient pressure, ΔP is the pressure overshoot, R is the universal gas constant, and T_f is the flame temperature. Substituting these into Eq. (1), and dividing by Eq. (2), a ratio of convective to conductive heat transfer yields Eq. (5), where $\bar{C}_{p,g}$ is the molar specific heat

$$v_g = \frac{-\kappa}{\phi \mu} \frac{\partial P}{\partial x} \sim \frac{-\kappa}{\phi \mu} \frac{\Delta P}{l_p}, \quad (3)$$

$$\bar{\rho}_g = \frac{(P_c + \Delta P)}{RT_f}, \quad (4)$$

$$\frac{q''_{conv}}{q''_{cond}} = \frac{(P_0 + \Delta P) \bar{C}_{p,g} \frac{\kappa}{\phi \mu} \Delta P}{RT_f k_{MIC}}. \quad (5)$$

This ratio is dependant on the pressure overshoot, ΔP , to the second power. It should be noted that the κ and ϕ are functions of the granular bed and not dependent on the pressurizing gas. For cases where the pressure overshoot is much greater than the chamber pressure, as in the fast-constant velocity regime, Eq. (5) suggests that the primary mechanism will be convection. In cases where the pressure overshoot is low or zero, Eq. (5) suggests that conductive heat transfer will be dominant.

Equation (5) is similar to the Peclet number, in that it is a dimensionless parameter relating

advection to diffusion. It is appropriate to acknowledge the Andreev number (An), which has been used in the literature to describe the transition from a conductive to a convective burn for reactive flows in porous media [20]. The Andreev number is defined as the product of the bulk density of the composite, the combustion wave speed, the hydraulic pore diameter, and the heat capacity of the composite divided by the thermal conductivity of the gas. It has been shown that for low values of An , slow burning was observed, with the conduction of heat dominating. Alternatively, for high values of An , a convective mechanism dominates flame propagation. A similar trend applies for Eq. (5).

For different pressurizing gases, the thermal conductivity of the nanoscale thermite will change, since the loosely packed thermite is about 90% interstitial gas. In the case of helium, the thermal conductivity of the nanoscale thermite will be enhanced compared to the cases of argon and nitrogen. This could be a possible explanation for why helium begins propagating in a slow-constant velocity mode at a much lower pressure than argon and nitrogen.

In the conductive regime, the propagation speeds are shown to slightly decrease with an increase in pressure. The speeds of the argon and helium systems match reasonably well, while the propagation speeds in nitrogen are slower. This suggests that the most important property in the conductive regime is the flame temperature, since increasing the pressure decreases the flame temperature, and due to its high specific heat, nitrogen will have a lower flame temperature than the other two pressurizing gases.

Although it is proposed that the propagation mechanism at high pressures is conductive and the results for the transition points in the three pressurizing gases seem to be explainable through this mechanism, further study is needed to substantiate this argument. For example, the role of radiative heat transfer in the absence of convection needs to be investigated. For the fast propagations, a simple order of magnitude analysis shows that buoyancy and free convection effects are negligible. However, for the slow propagations, these effects may begin to show some significance. Alternatively, the melt dispersion mechanism (MDM), which has been predicted by Levitas et al. [21], is another mode of propagation that may exist in the absence of convective burning. With this mechanism, when there is little gas production, as in the high-pressure cases, the advection of dispersed aluminum clusters would be the mode by which the reaction propagates the mixture. However, if this were the case, it is not apparent why the transition point to the slow constant velocity regime would have any dependence on the pressurizing gas.

The major conclusions of this work are that there exist multiple modes of propagation: constant velocity, accelerating, and oscillatory. This is indicative that the propagation mechanism is changing, likely from convective to a combination of convection and conduction, to a purely conductive propagation. Also, the different results observed with different pressurizing gases suggest that the heat transfer properties of the gas are important.

Acknowledgments

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